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The research goal is to further develop quantum well heterostructure (QWH) lasers and to realize reliable $Al_xGa_{1-x}As$-GaAs QWH lasers on Si. In spite of the significant lattice and thermal expansion mismatch between GaAs and Si, the idea of splicing III-V semiconductor technology, i.e., optoelectronics and photonics, onto Si has obvious appeal. Adding to this is the fact, as shown earlier in this work, that cw 300 K $Al_xGa_{1-x}As$-GaAs QWH lasers can be grown on Si, and that the Si substrate serves as a better heat sink than GaAs. This makes possible the "right-side-up" heat sinking needed for electronic-photonic integrated circuits.
This report contains research results on quantum well heterostructures on Si, impurity-induced layer disordering, phonon-assisted laser operations and other laser studies.


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# III-V SEMICONDUCTOR QUANTUM WELL LASERS AND RELATED OPTOELECTRONIC DEVICES ON SILICON

REPORT NO. 2


N. Holonyak, Jr./K. C. Hsieh/G. E. Stillman


December, 1989


U.S. ARMY RESEARCH OFFICE
DAAL 03-89-K-0008





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## I. INTRODUCTION

As reported previously our goal is to further develop quantum well heterostructure (QWH) lasers and to realize reliable $Al_xGa_{1-x}As$-GaAs QWH lasers on Si. In spite of the significant lattice and thermal expansion mismatch between GaAs and Si, the idea of splicing III-V semiconductor technology, i.e., optoelectronics and photonics, onto Si has obvious appeal. Adding to this is the fact, as shown earlier in this work, that cw 300 K $Al_xGa_{1-x}As$-GaAs QWH lasers can be grown on Si, and that the Si substrate serves as a better heat sink than GaAs. This makes possible the "right-side-up" heat sinking needed for electronic-photonic integrated circuits. It is interesting, however, that for over 2 1/2 years no one has reported cw 300 K laser operation of QWHs grown on Si that exceeds in operating lifetime the Urbana work of 1987 (4-24 hr cw 300 K laser operation).

The problem with the GaAs-Si mismatch, and the resulting defects, is sufficient to make unlikely anything but small steps in progress in QWH laser operation unless major new ideas are uncovered. Because these are apt to emerge slowly, we are concerned also with other QWH laser studies, with the goal in general to realize better lasers. These studies include impurity-induced layer disordering (IILD) of QWHs because of its fundamental importance, as well as its practical importance for lasers and other devices. Two further areas of IILD have emerged: (1) IILD via combined Si-O diffusion, and (1) IILD of $Al_yGa_{1-y}As$-GaAs QWHs in which a strained layer $In_xGa_{1-x}As$ secondary QW is inserted into the primary GaAs QW. An $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH is a particularly interesting compound QW structure because of its extended bandfilling range and its potential for extended broadband laser operation.

Another area of laser study receiving continued study is photopumping of QWHs because it permits identification of phonon-assisted recombination processes. In this work the importance of the semiconductor sample cavity Q, i.e., high Q vs. low Q, has emerged. The issue of devising more sophisticated semiconductor laser cavities is converging from phonon-assisted laser studies (Urbana) and from surface-emitting laser studies (of others), and suggests that more sophisticated cavities (not just cleaved end mirrors) will become common in semiconductor lasers.

## II. QUANTUM WELL HETEROSTRUCTURE ON Si

*Our early work (1986-87) has established that it is possible, in a two-*step MBE-MOCVD process, to grow cw 300 K (4-24 hr life) $Al_xGa_{1-x}As$-GaAs QWH lasers on Si. These results have not been duplicated in over 2 1/2 years and say something about the nature of the problem of building a high performance III-V injection device on Si. It is a simpler matter, however, to achieve short-lived QWH lasers on Si ($\lesssim$ 10 min). These results have been tabulated previously (Report No. 1, June, 1989) and are reproduced here, Table I, from W. E. Plano's Ph.D. thesis (Urbana, Ref. 1). Reference 1 shows that it is not very difficult to realize (via MBE-MOCVD) $\lesssim$ 10 min life QWH $Al_xGa_{1-x}As$-GaAs lasers on Si, but it is a far different matter to do better. Reference 1 shows also that $In_xGa_{1-x}As$ layers tend to bend threading dislocations, which is desired. On the other hand, it is not a very good idea to grow $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH lasers on Si, because of the tendency to increase the dislocation density in the device active region.

Besides the direct effect of lattice and thermal mismatch and defects (dislocations), there are other consequences of employing a Si substrate for $Al_xGa_{1-x}As$-GaAs QWH lasers. Defect-assisted auto-diffusion of substrate Si into the III-V QWH can occur at higher growth temperatures. This leads to undesired layer disordering, which obviously is a more serious problem at higher than lower QWH growth temperatures. We have described these effects in Ref. 2 and the appended abstract.

Finally, concerning QWHs grown on Si, it is worth noting that ultimately in some form of electronic-photonic integrated circuit it is not necessary to have continuous III-V semiconductor layers across the Si substrate. Some form of patterning will be required, and this is at once of interest in trying to terminate dislocations at structural edges or pattern edges. This work has not advanced as fast as desired because of down-time and overhaul of our Emcore 3000 DFM MOCVD reactor. We have, however, managed to make a start on this work by employing patterning by laser melting, followed by various annealing and diffusion operations. Our initial results are reported in Ref. 3 (and appended abstract).

Besides attempting to understand how to eliminate dislocations in QWHs grown on Si, we are studying the defect behavior of the best existing QWH lasers on Si at lower temperatures, where the operating life is improved. This work is in an early stage and will be reported later.

## III. IMPURITY-INDUCED LAYER DISORDERING

We know from previous work (Guido, et al., APL 52, 522 (1988)) that carbon (C) can conveniently be introduced by MOCVD as an acceptor in

$Al_xGa_{1-x}As$-GaAs QWHs. With a separate source ($CCl_4$) the C doping can be enhanced, and on a Column V lattice site (acceptor), serves as a convenient marker to study IILD. First, we note that C tends to be fixed against diffusion (Ref. 4). This is advantageous in fundamental studies of IILD because the usual Al-Ga-In atom interchange on the Column III lattice site can be determined to be dependent or independent of the atoms and processes occurring on Column V lattice sites. Some of our work in this area is now in the process of publication (Refs. 5,6,7).

Because of its practical importance, we are continuing work on IILD involving simultaneous Si-O diffusion in $Al_xGa_{1-x}As$-GaAs and $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWHs. Recent publications in this area are Refs. 8,9,10.

For other reasons, we have re-examined the earlier reports of others on the use of sulfur (S) for IILD. Curiously, S-IILD is now essentially dormant. Our data (Ref. 11) indicate that S, contrary to earlier reports, is not a very effective impurity for IILD, and in some respects may be as immobile as C. These results suggest why not much is happening with S-IILD; it is not a very effective method to produce IILD.

## IV. PHONON-ASSISTED LASER OPERATION

Because of its bearing on the fundamental aspects of carrier recombination and stimulated emission, and as a means to assess the quality of QWH material before painstaking device assembly, we continue to photopump QWH crystals and to study phonon-assisted laser operation. The extension of this work to $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWHs is described in Ref. 12. In Ref. 13 we describe this area of work more generally and review the importance of photopumping rectangular QWH samples (free of lower-gap substrate and contact layers) either with fully reflecting sample edges (high Q sample) versus natually reflecting cleaved edges (low Q sample). We have been able to devise a scheme, for use on a single QWH sample, of photopumping in either the low-Q or high high-Q sample configuration, and turning ON and OFF the laser operation that is possible on confined-particle transitions. This makes it possible to identify unambiguously the fact that the lower energy stimulated emission occurs shifted one phonon $\Delta\hbar\omega \approx 36$ meV $\approx \hbar\omega_{LO}$ below the confined-particle transitions. These results are described in Ref. 14.

V. OTHER LASER STUDIES

In addition to our interest in $Al_xGa_{1-x}As$-GaAs and $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWHs, we have continued work on the visible-spectrum system $In_{0.5}(Al_xGa_{1-x})_{0.5}P$-$In_{0.5}(Al_yGa_{1-y})_{0.5}P$. This is a difficult QWH system to master technologically (i.e., to control and to grow lattice matched free of defects). In spite of the fact that these crystals are harder to grow and are in poorer supply (Craford's group, H-P, San Jose), we are continuing these studies to extend and understand IILD more completely and to generate a visible-spectrum laser technology (see Ref. 15).

As previously mentioned (Report #1, June, 1989), $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWHs, with lower-gap InGaAs QWs within higher-gap GaAs QWs, present an opportunity for extended bandfilling and thus an uncommonly large laser tuning range in external grating cavities ($\Delta\hbar\omega \sim 150$ meV or $\Delta\lambda \sim 1000$ Å). These cw 300 K lasers are described in Ref. 16. We have received a number of inquiries about this work because of its obvious practical importance in showing that a semiconductor equivalent of a dye laser can be realized. We are extending this work to multiple-stripe lasers in an attempt to realize higher powers.

## VI. CONTRIBUTORS

The principal investigators contributing to various parts of the work reported here are:

1) N. Holonyak, Jr.
2) K. C. Hsieh
3) G. E. Stillman

(This report has been prepared by N. Holonyak, Jr.) The graduate students either receiving direct project support or otherwise contributing to various portions of the work reported here are:

1) J. M. Dallesasse, Ph.D. Student
2) D. C. Hall, Ph. D. Student
3) F. Kish, Ph.D. Student (A.T.&T. Fellowship)
4) J. S. Major, Jr., Ph. D. Student (Intel Fellowship, 1989-1990)
5) D. W. Nam, Ph. D. Student (Kodak Fellowship)
6) M. A. Plano (Stillman advisor, Ph.D. completed)
7) W. E. Plano (Ph.D. completed, now employed by Spectra Diode Laboratories, San Jose, California)
8) A. R. Sugg, Ph.D. Student

9) E. J. Vesely, Ph. D. Student (NSF Fellowship)

Note that some of the graduate students making contibutions to this work (Refs. 1-16) have received support from other projects or have received fellowship support. Another contributor, L. J. Guido, has concluded his post-doctoral appointment and has joined the EE Department of Yale University. Of the students listed above, Dallesasse, Hall, Major, and Nam are past their doctoral preliminary examinations, and Nam will finish his thesis in the Spring 1990 semester. We mention that the National Science Foundation Engineering Research Center has supported much of our MOCVD crystal growth (EMCORE reactor), and our NSF MRL has supported our TEM and SIMS analyses, which are spread throughout much of the work reported here.

We mention also that since impurity-induced layer disordering (IILD) originated in Urbana, the University of Illinois holds a number of fundamental patents on this technology. In 1989 the first license agreement with an industrial laboratory (Spectra Diode Laboratories) for commercial use of IILD patents was concluded.

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TABLE I

$Al_xGa_{1-x}As$-GaAs Quantum Well Heterostructure Laser Structures Grown by MOCVD on TI MBE GaAs-on-Si "Substrates."

| Crystal Number | Growth Temperature $T_g$ (°C) | Buffer Layers | Photopumped laser operation | Diodes | Comments |
|---|---|---|---|---|---|
| BP256 | 815 | AlGaAs-GaAs SL | pulsed | undoped | no QW |
| BP257 | 760-815-760 | AlGaAs-GaAs SL | pulsed | undoped | QW intact but poor light emitter |
| BP259 | 790 | AlGaAs-GaAs SL | pulsed | undoped | smeared QW |
| BP265 | 775 | AlGaAs-GaAs SL | cw died slowly | undoped | QW intact |
| BP287 | 775 | AlGaAs-GaAs SL | cw stable | undoped | Zn-diffused substrate |
| BP338 | 775 | AlGaAs-GaAs SL | --- | pulsed | high resistance |
| BP341 | 775 | AlGaAs-GaAs SL | --- | cw $\lesssim$ 10 min | $R_s \sim$ 15-20Ω |
| BP346 | 775 | AlGaAs-GaAs SL | --- | cw $\lesssim$ 10 min | $R_s \sim$ 15-20Ω |
| BP365 | 750 | AlGaAs-GaAs SL | pulsed | did not lase (DNL) | poor light emission |
| BP373 | 760 | AlGaAs-GaAs SL | --- | cw < 10 min | $R_s \sim$ 10-15 Ω |
| BP383 | 760 | AlGaAs-GaAs SL | cw | cw < 10 min | $R_s \sim$ 10-15 Ω 30 min anneal @ 760 before growth |
| BP426 | 760 | InGaAsP | cw stable | DNL | doping too low in InGaAsP (poor I-V's) |
| BP723 | 760 | InGaAs-GaAs SLS | pulsed | pulsed, died rapidly | high threshold low $R_s$ (~ 5 Ω) |
| BP761 | 760 | InGaAsP | pulsed | pulsed, died rapidly | high threshold low $R_s$ (~ 5 Ω) |
| BP762 | 760 | None | DNL | DNL | InGaAs QW |
| BP763 | 760 | 3 InGaAs wells | DNL | DNL | InGaAs QW |
| BP770 | 760 | 2 InGaAs wells | pulsed | pulsed, died | High threshold low $R_s$ (~ 5 Ω) |
| BP813 | 760 | 2 InGaAs wells patterned substrate | --- | pulsed, stable, DNL cw | High $R_s$ $R_s$ > 20 Ω |

GROWTH OF III-V SEMICONDUCTOR LASERS ON SILICON

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The growth of reliable III-V semiconductor lasers on Si would be a significant step toward the fabrication of an opto-electronic integrated circuit, but reliable III-V semiconductor lasers grown on Si have yet to be reported. Many problems not encountered when growing III-V lasers on III-V substrates must be overcome before III-V lasers grown on Si become reliable. In the experiments described here many different methods are used to try to improve the reliability of III-V lasers grown on Si.

Data are presented showing that impurity induced layer disordering (IILD) can be greatly accelerated when an abundance of dislocations are present. The high dislocation density of III-V materials grown on Si makes this a problem that cannot be overlooked. This problem can be minimized but it puts limits on the kind of thermal processing that can be done to a III-V laser grown on Si. Perhaps the most difficult problem encountered when growing III-V lasers on Si is dislocations. If the dislocation density can be reduced, then lasers grown on Si should become more reliable. Several methods of reducing the dislocation density of III-V lasers grown on Si have been tried. Data are presented showing that Zn diffusions, strained layer superlattices, and lattice matched buffer layers are all somewhat effective at lowering the dislocation density. The high dislocation density of III-V lasers grown on Si makes InGaAs strained layer active region lasers on Si an unlikely possibility because the dislocations tend to accumulate at the active region of these structures where dislocations are the most damaging.

Growth of III-V lasers on patterned GaAs-on-Si wafers has also been investigated. Growing over a selective area should reduce some strain and possibly lower the dislocation density. Data are presented showing that the dislocation density can be reduced by selective area epitaxy. But other problems arise when growing over a selective area and make device fabrication more difficult.

Ref. 2

# Dislocation-accelerated impurity-induced layer disordering of $Al_xGa_{1-x}As$-GaAs quantum well heterostructures grown on GaAs-on-Si

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Data are presented showing that dislocations and Si autodiffusion promote accelerated layer disordering of $Al_xGa_{1-x}As$-GaAs quantum well heterostructures grown on GaAs-on-Si "substrates" via metalorganic chemical vapor deposition. The accelerated impurity-induced layer disordering is more extreme at higher temperatures ( > 800 °C) and virtually nonexistent at lower temperatures ( $\lesssim$ 775 °C).

The growth of III-V laser structures on Si substrates has recently attracted much attention.[1-4] The major defect problems encountered in growing GaAs on Si result from the large ( ~4%) lattice mismatch between the two materials and the large difference in thermal expansion coefficients, which cause a high density of dislocations in the epitaxial GaAs. The dislocation density must be reduced before quantum well heterostructures (QWHs) on Si, specifically QWH lasers, become reliable devices. Various techniques have been tried to reduce the dislocations in GaAs-on-Si, including thermal annealing[5,6] and impurity diffusion.[7] Also at issue in this system is the stability of the heterointerfaces during subsequent high-temperature processing as required, for example, in fabricating buried-heterostructure lasers or in activating implants. In this letter we demonstrate dislocation-accelerated impurity-induced layer disordering[8] that occurs during Si autodiffusion in QWHs grown on Si.

The QWH laser crystals used in this work have been grown in a hybrid two-step process similar to previous work.[9] First, molecular beam epitaxy (MBE) is used to grow a 2 $\mu$m *n*-type GaAs layer ($n_{Si} \sim 10^{18}/cm^3$) on a (100) *n*-type Si substrate ($n_d \sim 10^{19}/cm^3$) tilted 3° toward the [011] direction.[10] The GaAs-on-Si wafer is then treated as a GaAs "substrate." The QWH laser crystals used throughout this work are grown by metalorganic chemical vapor deposition (MOCVD) in an Emcore GS 3000 DFM reactor on the GaAs-on-Si substrate. The MOCVD crystal layers consist of the following: (1) a 0.5 $\mu$m *n*-type GaAs buffer layer ($n_{Se} \sim 2\times10^{18}$), (2) a 0.15 $\mu$m *n*-type $Al_{0.15}Ga_{0.85}As$ buffer layer ($n_{Se} \sim 2\times10^{18}$), (3) an eight-period 0.25 $\mu$m *n*-type $Al_{0.40}Ga_{0.60}As$-GaAs superlattice (SL) region ($n_{Se} \sim 10^{18}$), (4) a 0.25 $\mu$m *n*-type $Al_{0.4}Ga_{0.6}As$ layer ($n_{Se} \sim 10^{18}$), (5) a 0.9 $\mu$m *n*-type $Al_{0.75}Ga_{0.25}As$ lower confining layer ($n_{Se} \sim 5\times10^{17}$), (6) a 0.2 $\mu$m undoped $Al_{0.20}Ga_{0.80}As$ waveguide region with a single 100 Å undoped GaAs QW at its center, (7) a 0.8 $\mu$m *p*-type $Al_{0.75}Ga_{0.25}As$ upper confining layer ($p_{Mg} \sim 10^{18}$), and (8) a 0.1 $\mu$m *p*-type GaAs ($p_{Mg} \sim 2\times10^{18}$) contact layer. This is essentially the same structure shown schematically in Ref. 2 (Fig. 1).

Figure 1 shows the laser spectra of two photopumped QWH laser crystals that have been grown simultaneously. The only difference is that (a) has been grown on a GaAs substrate ($n_{Si} \sim 2\times10^{18}/cm^3$, dislocation density $\lesssim 10^3/cm^2$) and (b) on a GaAs-on-Si wafer (dislocation density $10^7$–$10^8/cm^2$). The QWH crystals are grown at 825 °C at a growth rate of ~500 Å/min and a V/III ratio of ~80. The laser wavelength of (b) the QWH grown on GaAs-on-Si exhibits a large shift to higher energy. The transmission electron micrograph (TEM) of Fig. 2 shows that the QW in the case of the GaAs-on-Si crystal (b) is completely intermixed with the $Al_{0.20}Ga_{0.80}As$ waveguide region, e.g., as thoroughly as in conventional impurity-induced layer disordering (IILD).[8,11] Transmission electron microscopy shows that the SL buffer layers also are completely intermixed on the GaAs-on-Si crystal, while the QW and SL regions remain intact on the QWH grown on the GaAs substrate. The laser wavelength observed on the former [(b) in Fig. 1] corresponds to what would be expected from a double-heterostructure (DH) laser with an $Al_{0.20}Ga_{0.80}As$ active region, further confirming that the QW has been "absorbed."

Recent data show that impurity diffusion is accelerated in crystals that have a high defect and dislocation density.[12] In fact, for this case, the diffusion of Si through the QWH

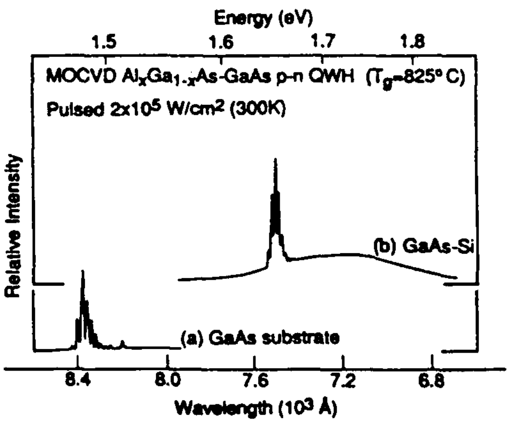


FIG. 1. Photoluminescence spectra of QWH lasers grown on (a) GaAs and (b) on GaAs-on-Si. The QWHs are grown simultaneously but the QWH laser grown on GaAs-on-Si operates (because of modification of the QW) at a much shorter wavelength.

  

# Defect-accelerated donor diffusion and layer intermixing of GaAs/AlAs superlattices on laser-patterned substrates

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Data are presented showing that donor diffusion and $Al_xGa_{1-x}As$-GaAs layer intermixing are greatly enhanced in the presence of defects created by crystal overgrowth on locally laser-melted substrates. Accelerated defect and impurity-induced layer disordering, and donor diffusion from a solid source ($SiO_2$), a vapor source (Ge), and from a grown-in source (Se) are observed in regions of high defect density. The enhanced donor diffusion and crystal self-diffusion are attributed to an increased density of column-III defects and dislocations in the crystal.

## I. INTRODUCTION

The discovery that impurity-induced layer disordering (IILD) intermixes $Al_xGa_{1-x}As$-GaAs heterostructures at temperatures much less than those required for thermal interdiffusion has led to many fundamental studies of crystal self-diffusion and impurity diffusion in III-V semiconductors.[1,2] Many studies point to the role of crystal defects in the atom diffusion and layer disordering processes.[2] These processes are found to be highly sensitive to the defect concentrations in the crystal. Impurity diffusion and IILD processes are generally governed by varying the defect concentrations in the crystal by controlling crystal surface conditions (e.g., encapsulants) and the annealing ambient. However, some heterostructures are grown with a large concentration of defects in the crystal (e.g., strained layer structures, quaternaries with imperfect lattice matching, and lattice mismatched epitaxial layers). These defects may alter the impurity diffusion and/or IILD processes. In this paper the role of increased crystal defect concentration on donor diffusion and layer intermixing is studied by deliberately creating regions of high defect density via a second crystal growth, overgrown crystal, on locally laser-melted substrates. Thermal annealing experiments with various encapsulants and donor diffusion from a solid source ($SiO_2$),[3] a vapor source (Ge),[4] and a grown-in source (Se)[5] show that accelerated impurity diffusion and layer intermixing result from an increased concentration of column-III defects and the dislocations in the crystal.

## II. EXPERIMENTAL PROCEDURES

The crystals for these experiments are grown by metalorganic chemical vapor deposition (MOCVD) in an Emcore GS-3000 DFM reactor. The Cr-doped semi-insulating GaAs substrates are first degreased and then etched in a 1:1 HCl:$H_2O$ mixture for 5 min. The substrates are rinsed with $H_2O$, blown dry with $N_2$, and loaded into the MOCVD reactor for the growth of the premelt layers. The initial layers, the premelt layers, are grown at 760 °C at a growth rate of 1000 Å/min, and consist of a 1000-Å GaAs buffer layer followed by a 3000-Å $Al_{0.75}Ga_{0.25}As$ layer. The $Al_{0.75}Ga_{0.25}As$ is then capped with a 2000-Å GaAs cap layer. All premelt layers are intentionally undoped. The GaAs cap layer prevents oxidation of the $Al_{0.75}Ga_{0.25}As$ during the subsequent laser patterning and enables high-quality, defect-free layers to be overgrown on the areas that are not laser patterned.

The laser patterning is performed by scanning an $Ar^+$ laser beam ($\lambda = 488$ nm) over the premelt layers so as to perform local laser melting in $\sim 4$-$\mu$m lines along the sample. The experimental arrangement is similar to that described elsewhere,[6] with a stage scan speed of 150 $\mu$m/s and an incident power level of $\sim 350$ mW. Single lines are prepared for transmission electron microscopy (TEM) and scanning electron microscopy (SEM) measurements. Six adjacent lines, with no spacing between stripes, are scanned to provide a broader laser-patterned region for secondary-ion mass spectroscopy (SIMS) measurements.

After the wafers are laser patterned, they are degreased again. The laser melting intermixes the GaAs cap layer with the underlying $Al_{0.75}Ga_{0.25}As$ layer, yielding $Al_xGa_{1-x}As$ ($0 < x < 0.75$) at the crystal surface. A $CF_4$ plasma is used to remove oxides instead of HCl, since HCl etches $Al_xGa_{1-x}As$. The wafer is placed in the $CF_4$ plasma at a power of 120 W for 5 min and is then loaded immediately into the MOCVD reactor for the second epitaxial crystal growth. These layers are grown at the same temperature and growth rate as the premelt epitaxial layers and consist of a 2000-Å GaAs buffer layer followed by a 2.0-$\mu$m 20-period 500-Å/500-Å AlAs-GaAs superlattice (SL), ending in a GaAs layer. The SL layers are grown either undoped or with the MOCVD reactor $H_2Se$ dopant flow adjusted to give $n_{Se} \sim 5 \times 10^{18}$ cm$^{-3}$ (as measured on a $C$–$V$ profiler). After the SL growth, the surface of the wafer is smooth and specular everywhere except where the laser patterning has been performed. The SL over the laser-patterned region is typically rough and cloudy in appearance, but still remains layered in form.

Before thermal annealing, the samples are rinsed in appropriate cleaning solvents and etched in $NH_4OH$ to remove any surface oxides. For Si diffusion into the crystal by $SiO_2$ reduction,[3] the top GaAs layer of the SL is removed with a clorox etch. Encapsulation layers of $Si_3N_4$ or $SiO_2$ ($\sim 1000$ Å) are grown on the crystal by chemical vapor deposition (CVD). Capless and encapsulated samples with $\sim 25$ mg of

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# Carbon diffusion in undoped, *n*-type, and *p*-type GaAs

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The effects of background doping, surface encapsulation, and $As_4$ overpressure on carbon diffusion have been studied by annealing samples with 1000 Å *p*-type carbon doping spikes grown within 1 $\mu$m layers of undoped ($n^-$), Se-doped ($n^+$), and Mg-doped ($p^+$) GaAs. The layers were grown by low-pressure metalorganic chemical vapor deposition using $CCl_4$ as the carbon doping source. Two different $As_4$ overpressure conditions were investigated: (1) the equilibrium $p_{As_4}$ over GaAs (no excess As), and (2) $p_{As_4} \sim 2.5$ atm. For each $As_4$ overpressure condition, both capless and $Si_3N_4$-capped samples of the $n^-$-, $n^+$-, and $p^+$-GaAs crystals were annealed simultaneously (825 °C, 24 h). Secondary-ion mass spectroscopy was used to measure the atomic carbon depth profiles. The carbon diffusion coefficient is always low, but depends on the background doping, being highest in Mg-doped ($p^+$) GaAs and lowest in Se-doped ($n^+$) GaAs. The influence of surface encapsulation ($Si_3N_4$) and $p_{As_4}$ on carbon diffusion is minimal.

In order to construct high-gain, high-frequency GaAs/AlGaAs heterojunction bipolar transistors (HBTs), it is necessary to grow a thin, heavily doped *p*-type base layer.[1,2] For GaAs/AlGaAs HBTs grown by metalorganic chemical vapor deposition (MOCVD), the acceptor impurities Mg and Zn are commonly employed in the GaAs base layer. Unfortunately, the large diffusion coefficients associated with these impurities lead to *p-n* junction redistribution, either during crystal growth itself or during subsequent high-temperature processing.[3–5] For example, the diffusion of Zn from the base into the *n*-type emitter changes the emitter-base *p-n* junction location relative to the emitter-base (GaAs/AlGaAs) heterojunction, thereby resulting in degraded HBT performance unless special set back doping layers are used.[5] Recent work has identified carbon as an alternative acceptor to Mg and Zn.[6–10] Because carbon incorporates primarily as a substitutional acceptor on the As sublattice (i.e., low interstitial carbon concentration), it is expected to have a much smaller diffusion coefficient than either Mg or Zn.[6,7,11,12] A detailed understanding of carbon diffusion in GaAs and AlGaAs would be useful for the prediction of the effects of high-temperature processing on HBT and other heterolayer device performance.

In the experiments described here, the effects of background doping, surface encapsulation, and arsenic overpressure ($p_{As_4}$) on carbon diffusion were studied by annealing (1000 Å) *p*-type carbon doping spikes grown in the center of 1 $\mu$m layers of undoped ($n^-$), Se-doped ($n^+$), and Mg-doped ($p^+$) GaAs. Secondary-ion mass spectroscopy (SIMS) was used to measure the carbon depth profiles for both as-grown and annealed samples. In general, the carbon diffusion coefficient is found to be extremely low, but depends on the background doping. The highest carbon diffusion coefficient is observed in Mg-doped ($p^+$) GaAs, while little or no carbon diffusion is found in Se-doped ($n^+$) GaAs. The effect of GaAs surface conditions was investigated by employing $Si_3N_4$ encapsulation and two different $As_4$ overpressures. These surface conditions had only a minor influence on the carbon diffusion coefficient. Estimates for the carbon diffusion coefficient in undoped ($n^-$) GaAs and Mg-doped ($p^+$) GaAs were found by fitting a one-dimensional diffusion equation solution to the measured SIMS carbon depth profiles.

The epitaxial layers used in this work were grown by low-pressure metalorganic chemical vapor deposition (MOCVD) in an Emcore GS3100 reactor on 2° off (100) oriented liquid-encapsulated Czochralski GaAs substrates. All growths were carried out at $T_G \sim 600$ °C, $P_G \sim 100$ Torr, substrate rotation rate $\sim 1500$ rpm, $H_2$ flow rate $\sim 9$ slm, growth rate $\sim 1000$ Å/min, and V/III ratio $\sim 60$. Trimethylgallium and 100% $AsH_3$ were the respective group III and V precursors. Nominally undoped GaAs grown under these conditions is *n*-type with a background carrier concentration of $n^- \sim 1 \times 10^{15}$ cm$^{-3}$. Layers doped with Mg (*p*-type) using $MCp_2Mg$ have a hole concentration of $p^+ \sim 1 \times 10^{19}$ cm$^{-3}$ as measured by electrochemical capacitance-voltage profiling. Layers doped with Se (*n*-type) using hydrogen selenide have an electron concentration of $n^+ \sim 5 \times 10^{18}$ cm$^{-3}$. All epilayers are $\sim 1$ $\mu$m in total thickness with the carbon doping spike (1000 Å) located 0.5 $\mu$m from the crystal surface. The carbon doping source, a 500 ppm mixture of $CCl_4$ in high-purity $H_2$, was turned on only during the growth of the carbon doping spike. The $CCl_4$ flow rate for each run was sufficient for the growth conditions used to achieve a carbon acceptor concentration of $p \sim 5 \times 10^{18}$ cm$^{-3}$.

All anneals were performed in evacuated quartz ampoules ($p \sim 10^{-6}$ Torr, vol $\sim 3$ cm$^{-3}$) at 825 °C for 24 h. For each ampoule the sample set (six samples) consisted of a capless and a $Si_3N_4$-capped sample of the three as-grown crystals ($n^-$, $n^+$, and $p^+$). Pyrolytic decomposition of $SiH_4$ and $NH_3$ at 700 °C was used to grow the 1000-Å-thick $Si_3N_4$ encapsulant. A large $As_4$ overpressure ($p_{As_4} \sim 2.5$ atm) was achieved by adding 25 mg of elemental As to the anneal

  

# COLUMN III-COLUMN V SUBLATTICE INTERACTION VIA Zn- AND Si-IMPURITY-INDUCED LAYER-DISORDERING OF $^{13}C$-DOPED $Al_xGa_{1-x}As$-GaAs SUPERLATTICES

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## ABSTRACT

Experiments are described employing secondary ion mass spectroscopy (SIMS) to study the stability of $^{13}C$-doped $Al_{0.5}Ga_{0.5}As$-GaAs superlattices against Zn- and Si-impurity-induced layer-disordering (IILD). The modulation depth of the SIMS $^{27}Al$ and $^{13}C$ signals is used as a sensitive probe of Column III and Column V sublattice interdiffusion. The data show that $C_{As}$ is much more stable against Zn- and Si-IILD than the Column III superlattice host crystal itself. The minor enhancement of $C_{As}$ diffusion via the Column III disordering agents, which is present to a significant extent for Si-IILD but almost non-existent for Zn-IILD, suggests that there is no direct interchange of Column III and Column V sublattice atoms. The Zn and Si enhancement of carbon diffusion is probably caused by local Coulombic interaction between the diffusing $Zn_i^+$ and $Si_{III}^+$ species and the $C_{As}^-$ acceptor.

# Al-Ga INTERDIFFUSION IN HEAVILY CARBON-DOPED $Al_xGa_{1-x}As$-GaAs QUANTUM WELL HETEROSTRUCTURES

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Impurity-induced layer disordering experiments on $Al_xGa_{1-x}As$-GaAs quantum well heterostructures (QWHs) that are doped heavily with carbon are described. The data show that carbon doping retards Al-Ga interdiffusion relative to an undoped crystal, and that interdiffusion in C-doped QWHs is not enhanced by a Ga-rich (vs. As-rich) annealing ambient. The data are inconsistent with most Fermi-level-effect models for layer disordering that do not include chemical impurity dependence or sublattice dependence, and that do not consider the possibility of inhibited Al-Ga interdiffusion in extrinsic crystals.

# IMPURITY-INDUCED LAYER DISORDERING: CURRENT UNDERSTANDING AND AREAS FOR FUTURE INVESTIGATION

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## ABSTRACT

The purpose of this work is to give an overview of the current phenomenological understanding of impurity-induced layer disordering (IILD). First, we identify key experimental findings such as the influence of the crystal surface-ambient interaction, the Fermi-level effect, and the impurity concentration on Al-Ga interdiffusion. Second, we review the strengths and weaknesses of existing IILD models in consideration of the above mentioned experimental data. Finally, we discuss the pitfalls involved in generalizing the results of individual Al-Ga interdiffusion experiments in order to explain a broader collection of IILD data.

## INTRODUCTION

Because of the importance of high-temperature processes in semiconductor device fabrication the self-diffusion of host crystal atoms, e.g., in bulk Ge [1] and GaAs [2], has been studied previously to better understand the mechanisms of impurity diffusion. The recent interest in self-diffusion or interdiffusion phenomena in III-V semiconductors has been stimulated by advances in the epitaxial growth of quantum well heterostructure (QWH) crystals. Growth techniques such as metal organic chemical vapor deposition allow the device designer to replace homojunctions with heterojunctions and thick active layers with quantum wells to improve device performance. In many cases the key to realizing improved device performance is to maintain the as-grown heterointerface abruptness during subsequent high-temperature processing.

The first study of Al-Ga interdiffusion by Chang and Koma [3] focused on the effects of high-temperature As-rich annealing on heterointerface abruptness. The experiment involved a series of GaAs-AlGaAs-GaAs sandwich structures with relatively thick ($\geq 1000$Å) undoped epitaxial layers. Post-annealing auger electron spectroscopy (AES) analysis showed that the Al-Ga interdiffusion coefficient ($D_{Al-Ga}$) is relatively small and

# High-power disorder-defined coupled stripe $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum well heterostructure lasers

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Data are presented describing continuous (cw) room-temperature laser operation of $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum well heterostructure (QWH) phase-locked arrays. The ten-stripe arrays have 3 $\mu$m emitters, with emitter to emitter spacing of 4 $\mu$m, and are patterned onto the QWH crystal using a self-aligned Si-O impurity-induced layer disordering (IILD) procedure. The IILD process is devised to provide limited layer intermixing to ensure optical coupling (across $\sim 1$ $\mu$m). The coupled stripe QWH lasers exhibit narrow twin-lobed far-field patterns that show unambiguously phase locking in the highest order supermode. The cw output power of the lasers (differential quantum efficiency 52%) is shown from threshold ($\sim 75$ mA) to over 280 mW (both facets, no optical coatings).

The useful wavelength of the $Al_yGa_{1-y}As$-GaAs heterosystem can be extended by incorporating a pseudomorphic $In_xGa_{1-x}As$ quantum well (QW) into the GaAs active region. Since the demonstration of continuous (cw) room-temperature (300 K) stimulated emission of various strained-layer $In_xGa_{1-x}As$ QWH crystals, with emission wavelengths as long as $\sim 1.1$ $\mu$m,[1,2] both material quality and device sophistication have improved. The devices fabricated include ridge-waveguide single stripe lasers,[3] high-power cw arrays,[4] and most recently low threshold buried heterostructure single-stripe lasers[5] fabricated via Si-O impurity-induced layer disordering (IILD).[5,6] The limitations of earlier devices are that index-guided single stripe lasers operate single mode and exhibit good spectral purity but are limited in output power, while high-power arrays lack the optical coupling between adjacent emitters that permits phase locking and allows single frequency operation. These limitations can be overcome with more refined methods of layer disordering than used in earlier IILD coupled stripe arrays.[7,8] In this letter we present data describing a ten-stripe phase-locked array delineated via a self-aligned Si-O IILD procedure.

The quantum well heterostructure (QWH) crystal used in this work has been grown via low-pressure metalorganic chemical vapor deposition (MOCVD).[4,5,9] The growth temperature is maintained at 800 °C except for the $In_xGa_{1-x}As$ QW, which is grown at 630 °C to prevent In desorption from the crystal surface. The epitaxial layers are grown on a Si-doped GaAs substrate in the following order: (1) a GaAs buffer layer, followed by (2) an *n*-type (Se) $Al_{0.8}Ga_{0.2}As$ lower confining layer ($\sim 1.25$ $\mu$m), (3) a nominally undoped waveguide (WG, Fig. 1) and active region, (4) a *p*-type (Mg) $Al_{0.8}Ga_{0.2}As$ upper confining layer ($\sim 6000$ Å), and (5) a *p*-type (Mg + Zn) GaAs contact layer ($\sim 1000$ Å). The active region is an $Al_{0.3}Ga_{0.7}As$ waveguide layer ($\sim 3000$ Å) that has in its center an $In_{0.1}Ga_{0.9}As$ QW ($\sim 100$ Å) within a thicker ($\sim 640$ Å) GaAs layer. The array fabrication procedure begins with the chemical vapor deposition of $\sim 1000$ Å of $Si_3N_4$. The $Si_3N_4$ is then patterned into a ten-stripe array with masking stripes of width 3 $\mu$m and stripe spacings of 4 $\mu$m center to center. With the photoresist still intact, the GaAs contact layer on the crystal is removed ($\sim 1000$ Å) between the stripes using a calibrated 1:8:80 ($H_2SO_4$:$H_2O_2$:$H_2O$) etch, thus leaving the high-gap $Al_{0.8}Ga_{0.2}As$ upper confining layer exposed. Next, 1400 Å of $SiO_2$ is electron beam evaporated onto the exposed high-gap material. This is followed by a lift-off procedure to remove both the photoresist and the $SiO_2$ from the $Si_3N_4$ stripes. The residual photoresist is removed by a 10 min $O_2$ plasma etch. The crystal, with $SiO_2$ left in the stripe openings, is then sealed in an evacuated quartz ampoule with no excess arsenic and is annealed at 825 °C for 12 h. The high-temperature anneal initiates Al reduction of the $SiO_2$ encapsulant, which is the source of the Si to perform the IILD

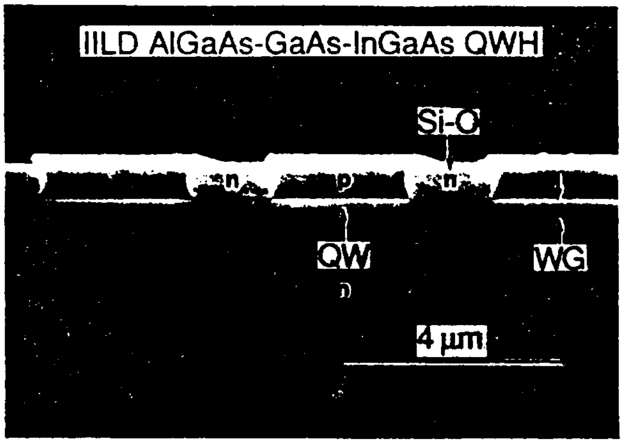


FIG. 1. Scanning electron microscope image of the cross section of an $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH crystal delineated via Si-O IILD. The 3 $\mu$m active region is protected by $Si_3N_4$ and remains intact throughout the IILD anneal. Between the stripes Al reduction of the $SiO_2$ encapsulant occurs at the high-gap $Al_{0.8}Ga_{0.2}As$ upper confining layer. The resultant Si-O diffusion moderately disorders the waveguide, allowing optical coupling between stripes.

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# High Performance $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ Quantum Well Lasers Defined by Silicon-Oxygen Impurity-Induced Layer Disordering

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In these experiments impurity-induced layer disordering (IILD) utilizing chemical reduction of $SiO_2$ by Al (from $Al_{0.8}Ga_{0.2}As$) is employed to generate Si and O to effect layer disordering. The $SiO_2$-$Al_{0.8}Ga_{0.2}As$ reaction is studied with respect to annealing ambient. By controlling the extent of disordering via $As_4$ overpressure, closely spaced ($\sim$1 $\mu$m) Si-O IILD buried heterostructure lasers can be optically coupled or uncoupled. Direct observation of O incorporation into the buried layers is shown using secondary ion mass spectroscopy (SIMS). The thermal stability of separate-confinement $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum well heterostructure (QWH) laser crystals is investigated using SIMS, transmission electron microscopy (TEM), and photoluminescence (PL) measurements. The data show that the thermal stability of a strained-layer $In_{0.1}Ga_{0.9}As$ quantum well (QW) is strongly dependent upon: (1) the layer thickness and heterointerfaces of the $Al_yGa_{1-y}As$-GaAs waveguide layers located directly above and below the QW, (2) the type of surface encapsulant employed, and (3) the annealing ambient. Narrow single-stripe (<2 $\mu$m) lasers fabricated via Si-O diffusion and layer disordering exhibit low threshold currents ($I_{th}$ $\sim$ 4 mA) and differential quantum efficiencies, $\eta$, of 22% per facet under continuous (cw) room-temperature operation.

**Key words:** InGaAs strained-layer laser, AlGaAs-GaAs-InGaAs quantum well heterostructure, impurity-induced layer disordering (IILD), Si-O IILD, coupled-stripe laser

## INTRODUCTION

Since the initial observation of continuous (cw) room-temperature (300 K) laser operation of strained-layer materials,[1] the study of pseudomorphic quantum well heterostructures (QWH's) for use in lasers and other optoelectronic devices has accelerated. Perhaps the most popular pseudomorphic heterosystem is simply that of a single $In_xGa_{1-x}As$ quantum well (QW) inserted into an $Al_yGa_{1-y}As$-GaAs QWH laser crystal. Strained layer lasers in the $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ system range from high power arrays, operating both with and without phasing-locking between elements, to low threshold single-stripe buried heterostructure laser diodes fabricated via Si-O impurity induced layer disordering (IILD).[2-4] This paper is concerned with the latter, specifically with two problems concerning buried heterostructures realized by Si-O IILD. The first is that of achieving a better understanding of the Al reduction of $SiO_2$ for co-diffusion of Si and O into the QWH crystal to effect Si-O IILD.[5,6] The role of $As_4$ overpressure during the anneal cycle and its effect on $SiO_2$ reduction is examined, with attention given to controlling the total amount of layer disordering by varying the $As_4$ overpressure. Second is that of determining the QWH parameters and conditions for a strained-layer heterostructure to be stable for extended annealing periods (*i.e.*, when masked against Si-O IILD) at temperatures exceeding 800° C. Although theoretical work exists concerning the stability of strained-layer $In_xGa_{1-x}As$ QWH's, these models are not specifically targeted at the annealing conditions required to fabricate IILD buried heterostructure devices. Recently it has been shown that the thermal stability of a pseudomorphic $In_xGa_{1-x}As$ QW, surrounded top and bottom by GaAs, is improved by increasing the thickness of the GaAs overlayer from 200 to 5000Å.[7] In this paper we consider the possibility that heterointerfaces within $\sim$500Å of an $In_xGa_{1-x}As$ QW (buried beneath $\sim$7000Å of $Al_xGa_{1-x}As$) have a major influence on the thermal stability of the strained-layer QW in an $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH. In addition, by depositing different wafer encapsulants ($Si_3N_4$ or $SiO_2$) onto these crystals, we are able to alter the thermal stability of the strained-layer $In_xGa_{1-x}As$ QW. It is known that these two encapsulants ($Si_3N_4$ or $SiO_2$) create different populations of native defects that diffuse into the crystal and

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# COLUMN III VACANCY-INDUCED AND IMPURITY-INDUCED LAYER DISORDERING OF $Al_xGa_{1-x}As$-GaAs HETEROSTRUCTURES WITH $SiO_2$ OR $Si_3N_4$ DIFFUSION SOURCES

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## ABSTRACT

Experiments are described determining the critical parameters for vacancy- and impurity-induced layer disordering (IILD) of $Al_xGa_{1-x}As$-GaAs quantum well heterostructure (QWH) crystals that utilize $SiO_2$ and $Si_3N_4$ diffusion source layers. The $SiO_2$- or $Si_3N_4$-capped QWH crystal surface reaches equilibrium with the external annealing ambient by diffusion of Ga and As through the encapsulant layer, thereby determining the crystal surface deviation from stoichiometry and the Column III vacancy concentration for layer disordering. By proper design of the QWH crystal, encapsulant layer thickness, and annealing ambient the $SiO_2$ ($Si_3N_4$) can be employed as a Column III vacancy source (or mask) or as a Si and O (or N) diffusion source.

# LAYER DISORDERING OF n-TYPE (Se) AND p-TYPE (C) $Al_xGa_{1-x}As$-GaAs SUPERLATTICES BY S DIFFUSION

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Data are presented showing limited layer disordering of S-diffused Se-doped or C-doped $Al_xGa_{1-x}As$-GaAs superlattices. The S diffusion is characterized via secondary ion mass spectroscopy, shallow angle beveled cross sections, and absorption measurements. Limited layer disordering of Column-III-site atoms (Al $\rightleftarrows$ Ga) as well as minimal displacement of the Column-V-site acceptor C are observed. The S diffusion depth is much greater than that of the layer disordering, the magnitude of which is comparable with that of native-defect vacancy-assisted disordering (vacancy, $V_{III}$).

# Phonon-assisted stimulated emission in strained-layer $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum-well heterostructures

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Data are presented demonstrating phonon-assisted laser operation (77 and 300 K) of photopumped strained-layer $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ ($x \sim 0.15$) quantum-well heterostructures (QWHs) grown using metalorganic chemical vapor deposition. When a cleaved rectangular sample (10–50 $\mu$m × 100–500 $\mu$m) of the QWH, with GaAs substrate removed, is imbedded in In under a sapphire window (for 77-K data), and the In is folded upward along the cleaved edges to provide high edge reflection and high cavity $Q$, closely spaced end-to-end laser modes (9000 Å) occur along the sample at an energy one LO phonon below the lowest confined-particle transition ($\Delta E = \hbar\omega_{LO} \simeq 36$ meV), and widely spaced edge-to-edge laser modes occur across the sample on confined-particle transitions. For comparison, the experiment is repeated with rectangular QWH samples clamped on Au with a sapphire window, but with no metal folded onto the sample edges, thus insuring low reflectivity at the cleaved edges (low $Q$ cavity). In the low $Q$ resonator configuration, all of the high-energy modes (transitions on confined-particle states) disappear, and only the low-energy phonon-assisted laser modes are evident. This comparison (high $Q$ versus low $Q$ photoexcitation), as well as the abrupt turn-on of laser operation in a narrow spectral range one phonon ($\Delta E = \hbar\omega_{LO} \simeq 36$ meV) below the lowest confined-particle transitions, leads to unambiguous identification of phonon-assisted laser operation of a strained-layer $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH. In addition, bandfilling is demonstrated through the entire well depth of an $L_z \simeq 125$ Å $In_xGa_{1-x}As$ QW to well above 150 meV into the GaAs QW containing the strained layer.

## I. INTRODUCTION

Strained-layer $In_xGa_{1-x}As$ grown epitaxially on GaAs or $Al_yGa_{1-y}As$ has been studied extensively recently as a possible candidate for long wavelength optical devices as well as for its unique optical[1] and electrical[2] properties. The successful demonstration of continuous (cw) room-temperature (300 K) laser operation of strained-layer GaAs-$In_xGa_{1-x}As$ superlattices (SLs) and quantum-well heterostructures (QWHs)[3,4] has led to increased research aimed at improving strained-layer QWH lasers incorporating $In_xGa_{1-x}As$ active layers. As a result a number of workers have reported high-quality long-wavelength ($\lambda \sim 1$ $\mu$m) $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH lasers,[5,6] including disordered-defined low threshold stripe-geometry strained-layer QWH lasers ($\lambda \sim 910$ nm).[7] In addition, many Raman scattering experiments have led to identification of GaAs-like LO phonons ($36 \rightarrow 35$ meV, $x = 0 \rightarrow 0.15$) and InAs-like TO phonons in $In_xGa_{1-x}As$ epitaxial layers[8] and GaAs-$In_xGa_{1-x}As$ SLs.[9,10] However, stimulated emission in strained-layer $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWHs involving phonon-assisted processes has largely been ignored or simply has not been identified, which is the subject of this work.

A schematic diagram showing the downward scattering of electrons in the step density of states of a QWH, and for contrast in the constraint of a parabolic density of states corresponding to a bulk sample (dashed lines), is shown in Fig. 1. As the photogenerated hot electrons enter the recombination region (QW), the electrons thermalize downward producing phonons. Once an electron reaches the bottom confined-particle state, it can either recombine via a confined-particle transition ($n = 1$ electron-to-heavy hole, $n = 1\, e \rightarrow hh$; $n' = 1'$ electron-to-light hole, $n' = 1'\, e \rightarrow lh$) or $\Delta E = \hbar\omega_{LO}$ lower in energy via a phonon-assisted transition.

Since the first reports of the phonon-assisted photopumped laser operation of single and multiple-quantum-well $Al_xGa_{1-x}As$-GaAs heterostructures grown via metalorganic chemical vapor deposition (MOCVD),[11–14] several theories have been offered to explain,[15] and other arguments to dispute,[16–20] the existence of phonon-assisted laser operation. There is no indication, however, that the negative experiments, based mainly on QWHs and SLs grown by molecular-beam epitaxy (MBE), have been performed on cleaved samples with the substrates removed and with the (100) QWH samples (rectangles) heat sunk imbedded in In.[21] As we have recently shown,[22] not only does this result in excellent heat sinking for high level photoexcitation, but more important, it allows In reflectors to be folded up along the

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TTL03

SEN03 1 **Photopumping of Quantum Well Heterostructures at High or Low $Q$: Phonon-Assisted**
14 **Laser Operation**

ADR03
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ABS03
SEN03 1 Data are presented showing the basic difference in the stimulated emission spectrum of a photopumped $Al_xGa_{1-x}As$–GaAs
18 or $Al_yGa_{1-y}As$–GaAs–$In_xGa_{1-x}As$ quantum well heterostructure (QWH) heat sunk in a high-$Q$ versus a low-$Q$ cavity
SEN06 33 configuration. In the high-$Q$ case a 1–2 μm thick narrow (25–50 μm) cleaved rectangle, with the (100) GaAs substrate
20 removed, is heat sunk compressed in In under a sapphire, giving a high cavity photon lifetime because of metal reflectors
SEN09 40 folded up along the four samples edges. In the latter case (low $Q$) the (100) QWH rectangle is clamped under a sapphire
17 into simple contact with Au, leaving the four cleaved {110} sample edges lossy and yielding, compared to carrier thermalization
SEN12 36 times, a short resonator photon lifetime across the sample. For photopumping (77 K, $Ar^+$ laser, 5145 Å) of a low-$Q$ QWH
14 sample, only lower energy recombination radiation is observed, including phonon-assisted laser operation (provided that the
29 QWH is designed with good carrier, phonon, and photon confinement and with low alloy composition $Al_xGa_{1-x}As$ thermalization
SEN15 46 layers generating GaAs-like phonons near the QW). For photopumping of an otherwise similar QWH heat sunk in the high-$Q$
14 configuration (long photon lifetime across the sample), recombination at higher energy can compete with carrier thermalization,
30 and laser operation is observed on the confined-particle transitions, thus making unambiguous the identification of phonon
SEN18 46 sideband laser operation. Comparison of various QWHs heat sunk in the form of low-$Q$ or high-$Q$ resonators reveals the
18 heterostructure layer configurations appropriate for phonon-assisted laser operation.

TXT03
SEN03 1 ***Introduction***
PAR03
SEN03 1 Ever since the transistor,[1] interest in semiconductors and their FNT 1
SEN06 11 optical properties has grown. Fundamental to a semiconductor
6 is its energy gap, an electron–hole energy gap that ensures in-
SEN09 16 teresting optical properties. We mention that in the "beginning"
8 it was *not* known whether Ge (the first transistor material) was
19 direct gap or, as it happened to be, indirect gap,[2] which, indeed, FNT 2
31 proved to be vital for the relatively long carrier lifetimes making
SEN12 42 possible the first bipolar transistors.[1] It is interesting to note that
8 the distinction between a direct gap and an indirect energy gap
19 was not lost on some of the earlier workers who considered using
31 recombination radiation to achieve stimulated emission in a
SEN15 39 semiconductor.[3,4] The importance of this for semiconductor lasers FNT 3,4
SEN18 9 became quite apparent later.[5-8] Immediately with the synthesis FNT 5–8
6 and discovery of the semiconductor behavior of III–V compounds,[9] FNT 9
15 direct-gap materials became available, and thus, if not initially
24 that different in electrical properties than, say, Ge, they indeed
SEN21 34 proved to be different in optical properties. It is the direct-bandgap
6 III–V compounds that lie at the heart of optoelectronics and,
16 because they support stimulated emission (stimulated recombi-
22 nation radiation), are of concern here.
PAR06
SEN03 1 The difference between direct- and indirect-gap semiconductors
9 is apparent in absorption measurements, and even more so in
19 photoluminescence measurements, which, indeed, prove to be one
27 of the simpler and more convenient methods of assessing semi-
SEN06 36 conductor optical properties. This is particularly important for
7 III–V semiconductors because of their strong light-emitting
14 properties,[10] not to mention the fact that they serve as the basis FNT 10
SEN09 26 for heterojunctions and quantum well heterostructures. Photo-
2 luminescence, or at higher levels, photopumping, has the advantage
11 that experimental samples may be homogeneous or of the form
21 of sophisticated heterostructures or may be doped or undoped.
SEN12 1 The latter is important in fundamental studies where the com-
SEN15 11 plications of doping impurities are not desired. In addition,
4 photopumping has the further advantage that, without the need

JP

# Variable resonator (variable $Q$) photopumped phonon-assisted quantum well laser operation

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Data are presented on a photopumped rectangular ($w = 40\,\mu$m, $l = 250\,\mu$m) quantum well heterostructure (QWH), with variable resonator $Q$ along the sample, showing that phonon-assisted laser operation predominates. To achieve variable resonator $Q$, the QWH sample is heat sunk embedded in In over half of its length (reflecting edges, high $Q$), as opposed to simple contact with a Au shim over the remaining half and no reflecting metal on the sample edges (low $Q$). Photopumping at the low $Q$, high $Q$ boundary near the sample center turns on and off the $n = 1$ confined-particle recombination transition ($E_{11}$) and sets a higher energy experimental (as well as calculated) reference, $\hbar\omega_2 = E_{11}$, for the lower energy phonon-assisted laser operation, $\hbar\omega_1 = E_{11} - \hbar\omega_{\rm LO}$.

In recent work[1-4] we have shown that the issue of whether phonon-assisted laser operation of photopumped quantum well heterostructures (QWHs) can be observed unambiguously is not a matter of the samples being grown by metalorganic chemical vapor deposition (MOCVD)[5] or by molecular beam epitaxy (MBE),[6] but rather how the samples are prepared and heat sunk, whether as high or low $Q$ resonators. For a typical rectangular cleaved (100) QWH sample, it matters a great deal for the cavity photon lifetime (along or across the sample) if the crystal is free of absorptive lower gap contact layers and substrates, and if the {110} cleaved edges are left naturally reflecting ($R \sim 0.32$, low $Q$) or are covered with metal and are "fully" reflecting ($R \sim 0.9$–$0.99$, high $Q$). We recall that the cavity photon lifetime $t_c$ (no absorption) is

$$\frac{1}{t_c} = \frac{2c}{\eta}\frac{1}{2w}\ln\left(\frac{1}{R_1R_2}\right), \tag{1}$$

where $c$ is the speed of light, $\eta$ is the sample index of refraction ($\sim 3.6$), and $w$ (or $l$) is the sample width (or length). For example, for a $w = 40$-$\mu$m-wide QWH as in Fig. 1 ($l = 250\,\mu$m), the edge-to-edge cavity lifetime $t_c$ across the sample shifts, low $Q$ to high $Q$, from $t_c \sim 2.1\times10^{-13}$ s ($R \sim 0.32$) to $t_c \sim 2.3\times10^{-12}$–$2.4\times10^{-11}$ s ($R \sim 0.9$–$0.99$).[4] That is, the cavity photon lifetime across the sample shifts from less than to greater than the typical hot-electron thermalization time of $\sim 10^{-12}$ s (photopumped QWH, Ar$^+$ laser, 2.41 eV). Thus, in the high $Q$ case and longer photon lifetime, stimulated recombination can compete with carrier scattering. This makes possible higher energy laser operation across the fully pumped width of the sample on the confined-particle transitions. This form of laser operation is characterized by widely spaced modes ($\Delta\lambda_w \equiv \Delta\lambda_2 = 24$ Å) and serves as an important experimental reference, i.e., hot-carrier recombination at higher energies on confined-particle transitions and not downward scattering and recombination at the lowest possible transitions. In contrast, for low cavity $Q$ across the sample ($t_c < 10^{-12}$ s), stimulated emission is favored at the higher $Q$ that exists along the sample ($l > w$, closely spaced modes, $\Delta\lambda_l \equiv \Delta\lambda_1 = 4.6$ Å). This occurs on the lower energy processes that might exist below the fundamental absorption, below the confined-particle transitions, e.g., a many-particle-shifted or phonon-assisted process. In this letter we show, on a single QWH sample of variable $Q$ by choice of pump position and by homogeneous photopumping across the sample and inhomogeneous pumping along its length, that phonon-assisted laser operation predominates (cf. Fig. 1 of Ref. 3).

For these experiments we employ $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ ($x \sim 0.15$) QWHs that are grown by MOCVD

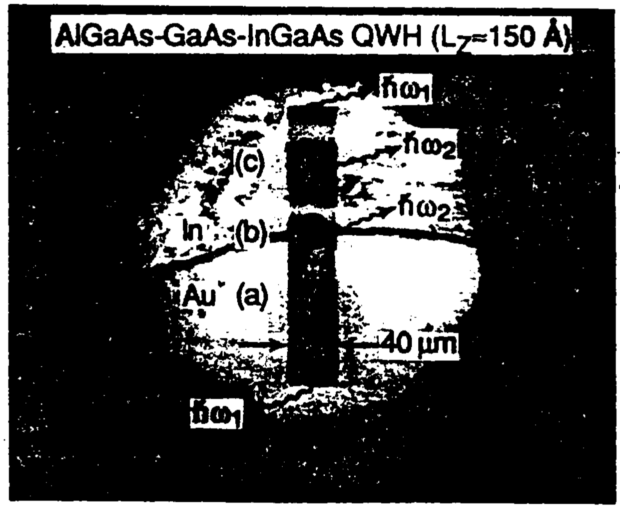


FIG. 1. Cleaved (100) $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ ($x \sim 0.15$) quantum well heterostructure (QWH) sample ($40\,\mu$m $\times$ $250\,\mu$m) embedded in In at the top (high $Q$) and in simple contact with Au at the bottom (low $Q$). (a) Photopumping and low $Q$ laser operation $\hbar\omega_1$ on closely spaced modes ($\Delta\lambda_l = \Delta\lambda_1 = 4.6$ Å) along the sample. (b) Photopumping at the low $Q$, high $Q$ Au-In boundary with laser operation on $\hbar\omega_1$ along and $\hbar\omega_2$ across the sample ($\Delta\lambda_w = \Delta\lambda_2 = 24$ Å). (c) Photopumping and high $Q$ laser operation $\hbar\omega_2$ across and $\hbar\omega_1$ along the sample ($\Delta\hbar\omega \equiv \hbar\omega_2 - \hbar\omega_1 = \hbar\omega_{\rm LO}$, where $\hbar\omega_2 = E_{11}$ is the $n = 1$ electron-to-heavy hole confined-particle transition).

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[b] National Science Foundation Doctoral Fellow.

  

# Impurity-induced layer disordering in $In_{0.5}(Al_xGa_{1-x})_{0.5}P$-InGaP quantum-well heterostructures: Visible-spectrum-buried heterostructure lasers

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Diffusion of Si into quantum-well heterostructures and superlattices employing the high gap III-V quaternary system $In_y(Al_xGa_{1-x})_{1-y}P$ is shown to result in impurity-induced layer disordering. Secondary ion mass spectroscopy, transmission electron microscopy, and photoluminescence data indicate that the diffusion of Si into an InAlP-InGaP superlattice grown lattice matched on GaAs ($y \approx 0.5$) results in the intermixing of the layers, thus forming an alloy of average composition. Buried-heterostructure lasers are fabricated using Si layer disordering of $In_{0.5}(Al_xGa_{1-x})_{0.5}P$ *p-n* quantum-well heterostructures. The disorder-defined stripe-geometry diode lasers operate pulsed at 300 K near 6400 Å. Continuous wave operation at $\lambda \sim 6255$ Å is achieved at $-47$ °C.

## I. INTRODUCTION

One of the more interesting developments in the study of quantum-well heterostructures (QWHs) and QW lasers has been the discovery that the diffusion of impurities into a III-V heterostructure (a layered crystal) enhances intermixing of the individual layers. Impurity-induced layer disordering (IILD) has been studied extensively in the $Al_xGa_{1-x}As$-GaAs system,[1-7] where it has been used to produce buried-heterostructure semiconductor lasers and laser arrays with superior performance characteristics.[8-11] As is well known, buried-heterostructure stripe-geometry lasers outperform other stripe geometries because of superior waveguiding and current confinement,[12] which in the present case is afforded by higher gap intermixed layers. The study of IILD in $Al_xGa_{1-x}As$-GaAs and other heterosystems is of fundamental importance, and is specifically of some importance in producing high-performance semiconductor lasers, and in general for use in optoelectronics. (See Ref. 13 for a review of IILD and its applications.)

An important III-V heterosystem for use in visible-spectrum lasers and to consider as a candidate for IILD is the quaternary $In_y(Al_xGa_{1-x})_{1-y}P$. This quaternary, which is a modification of the high gap ternary $In_yGa_{1-y}P$,[14] is of specific interest because of its known high direct–indirect crossover,[15] as well as the fact that IILD works particularly well for the case of Al-Ga substitution. Thus far, only a few IILD experiments have been performed in this heterosystem.[16] Through the use of metalorganic chemical vapor deposition (MOCVD),[17] it is possible to produce high-quality stacked layers (quaternary or ternary layers) of different Al and Ga composition, and thus of different energy gap. By analogy with the $Al_xGa_{1-x}As$ system, this makes possible the growth of heterostructures suitable for semiconductor lasers in the visible portion of the spectrum.[18-23] For example, room-temperature continuous (cw) operation of *p-n* diode $In_{0.5}(Al_xGa_{1-x})_{0.5}P$ QW lasers has already been demonstrated at wavelengths as short as 6395 Å.[24,25] Continuous (cw) photopumped laser operation at 300 K has been achieved at wavelengths as short as 6250 Å.[26,27] Because of the short wavelength capability of the $In_y(Al_xGa_{1-x})_{1-y}P$ system, the possibility of producing buried-heterostructure *p-n* diode QW lasers by IILD in this III-V material is of considerable interest. Previously, Zn diffusion in an $In_{0.5}(Al_xGa_{1-x})_{0.5}P$-GaAs heterostructure has been demonstrated to result in IILD, but has not been used to fabricate buried-heterostructure devices.[16] In the present work Si diffusion is demonstrated to result in the intermixing of $In_{0.5}(Al_xGa_{1-x})_{0.5}P$ layers grown lattice matched on GaAs. Silicon-IILD is used to produce buried-heterostructure QW diode lasers.

## II. LAYER DISORDERING IN THE $In_{0.5}(Al_xGa_{1-x})_{0.5}P$ SYSTEM

### A. Experimental procedure

In order to study the properties of Si IILD in the $In_{0.5}(Al_xGa_{1-x})_{0.5}P$ system, Si diffusion has been performed into an undoped 20-period $In_{0.5}Al_{0.5}P$-$In_{0.5}Ga_{0.5}P$ superlattice (SL, $L_B \approx 210$ Å, $L_z \approx 200$ Å). The ternary SL is grown by low-pressure (100-Torr) MOCVD on a (100) *n*-type GaAs substrate in an EMCORE GS3000-DFM reactor. Sources of the various crystal constituents are trimethylindium (TMIn), trimethylaluminum (TMAl), triethylgallium (TEGa), and phosphine ($PH_3$). The crystal growth parameters are chosen so as to minimize lattice mismatch between the $In_y(Al_xGa_{1-x})_{1-y}P$ layers and the GaAs substrate. The degree of lattice match is assessed by rotating crystal x-ray diffractometry on thick test layers of fixed com-

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# Broadband long-wavelength operation (9700 Å $\gtrsim \lambda \gtrsim$ 8700 Å) of $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum well heterostructure lasers in an external grating cavity

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Data are presented on *p-n* $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ quantum well heterostructure lasers showing that the large band filling range of a combined GaAs-$In_xGa_{1-x}As$ quantum well makes possible a very large tuning range in external grating operation. Continuous 300 K laser operation is demonstrated in the 8696–9711 Å range ($\Delta\lambda \sim 1000$ Å, $\Delta\hbar\omega \sim 150$ meV) and pulsed operation in the 8450–9756 Å range ($\Delta\lambda \sim 1300$ Å, $\Delta\hbar\omega \sim 200$ meV). The band filling and gain profile are shown to be continuous from the $In_xGa_{1-x}As$ quantum well ($L_z \sim 125$ Å, $x \sim 0.2$) up into the surrounding GaAs quantum well ($L_z \sim 430$ Å).

It has been previously demonstrated[1,2] that $Al_xGa_{1-x}As$-GaAs quantum well heterostructure (QWH) diode lasers can be operated in an external grating cavity over broadly tunable wavelength ranges. Continuous (cw) room-temperature tunable operation has been reported with a spectral range of $\Delta\hbar\omega \sim 100$ meV (8300 $\gtrsim \lambda \gtrsim$ 7800 Å) for single-stripe single-well AlGaAs QWH lasers,[1] and of $\Delta\hbar\omega \sim 94$ meV (8085 Å $> \lambda >$ 7620 Å) for high output power ($\sim 75$ mW) multiple-stripe, multiple-well AlGaAs QWH lasers.[2] Recently pulsed laser operation has been reported[3] for a single-well AlGaAs graded-index separate confinement heterostructure (GRINSCH) laser over the tuning range of 8500–7630 Å ($\Delta E \sim 166$ meV). It is possible to extend the useful wavelength range of the $Al_yGa_{1-y}As$-GaAs material system, and potentially its tuning range, by incorporating into it pseudomorphic $In_xGa_{1-x}As$ quantum wells. Since the earlier demonstration of continuous 300 K stimulated emission in GaAs-$In_xGa_{1-x}As$ strained-layer superlattices[4] and QWH lasers ($\lambda \lesssim 1.1$ $\mu$m),[5] there have been several reports of high quality strained layer $Al_yGa_{1-y}$ As-GaAs-$In_xGa_{1-x}As$ QWH *p-n* diode lasers.[6-10] This form of QWH diode laser makes possible a greater range of band filling (with less absorption) and thus provides an opportunity for a greater tuning range, which is the subject of this letter. Continuous (cw) 300 K tunable laser operation of an $Al_yGa_{1-y}As$-GaAs-$In_xGa_{1-x}As$ QWH is demonstrated in a $\Delta\hbar\omega \sim 150$ meV spectral range (9711 Å $> \lambda >$ 8696 Å). Pulsed laser operation (300 K) is demonstrated over a $\Delta\hbar\omega \sim 200$ meV tuning range (9756 Å $> \lambda >$ 8450 Å). The band filling and gain profile are shown to be continuous from the $In_xGa_{1-x}As$ QW ($L_z \sim 125$ Å, $x \sim 0.2$) up into the enclosing GaAs QW ($L_z \sim 430$ Å).

The strained-layer QWH crystal used in this work is grown by low-pressure metalorganic chemical vapor deposition (MOCVD).[7-9,11] The structure is grown on a Si-doped GaAs substrate, and consists of (1) an *n*-type (Se-doped) GaAs buffer layer, (2) an *n*-type (Se-doped) $Al_{0.8}Ga_{0.2}As$ lower confining layer ($\sim 1.35$ $\mu$m), (3) a nominally undoped waveguide and QW active region, (4) a *p*-type (Mg-doped) $Al_{0.8}Ga_{0.2}As$ upper confining layer ($\sim 1.15$ $\mu$m), and (5) a *p*-type (Mg- and Zn-doped) GaAs contact layer ($\sim 1800$ Å). The double-well active region is enclosed in an $Al_{0.35}Ga_{0.65}As$ waveguide ($\sim 0.5$ $\mu$m) and consists of an $In_xGa_{1-x}As$ QW ($L_z \sim 125$ Å, $x \sim 0.2$) within a GaAs QW ($\sim 430$ Å). The entire structure is grown at 800 °C except for the $In_xGa_{1-x}As$ QW which is grown at 630 °C to prevent the desorption of In from the crystal surface. A transmission electron microscope (TEM) image of the crystal is shown elsewhere (Fig. 7 of Ref. 12).

Stripe geometry laser diodes are fabricated from this QWH crystal by photolithographically defining 10 $\mu$m openings in a $SiO_2$ layer ($\sim 1000$ Å) grown by chemical vapor deposition (CVD). After thinning the crystal to a thickness of $\sim 105$ $\mu$m and metallizing the contacts, the wafer is cleaved into bars. For this work, a 150 $\mu$m bar is coated with a $\lambda/4$ $Al_2O_3$ antireflecting (AR) coating ($\lambda \sim 9200$ Å, $R \lesssim 2\%$). The back facet is coated with a high-reflectance (HR) dielectric mirror ($Al_2O_3$/Si). After sawing into individual die, the diodes are mounted *p* side down with In on Cu heat sinks.

The external grating cavity consists of an $f/1.0$ 50.8 mm lens to collimate the diode emission and a ruled diffraction grating used in the Littrow configuration. A small portion of the collimated beam is diverted to a 0.5 m monochromator equipped with an *S* 1 response photomultiplier. For cw operation, the diode temperature is stabilized at room temperature with a thermoelectric cooler.

Figure 1(a) shows a spontaneous emission spectrum (no feedback) for one of the diodes described above at the low injection level of 2 mA cw. The large tail extending toward higher energy in Fig. 1(a) shows that injected carriers fill the $In_xGa_{1-x}As$ QW up to the GaAs band edge ($\lambda \sim 8700$ Å, 300 K). At higher injection levels, band filling and the kinetics of carrier recombination cause the spontaneous emission peak to shift towards higher energy. Figure

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